Selective growth of InGaN quantum dot structures and their microphotoluminescence at room temperature

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We have fabricated InGaN quantum dot (QD) structures on hexagonal pyramids of GaN, using metalorganic chemical vapor deposition with selective growth. Intense photoluminescence was observed from the sample at room temperature. To directly observe the emitting areas, microphotoluminescence intensity images with a spatial resolution of a few hundred nanometers were used. The images show the emission was only from the tops of the hexagonal pyramids. The width of the emitting areas is about 300 nm, which is comparable to the spatial resolution of the images. Such a narrow width of emission areas indicates that InGaN QDs are formed on the tops of pyramids. © *2000 American Institute of Physics.* [S0003-6951(00)01022-6]

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GaN and related materials have been studied intensively, because of their usefulness for optical devices emitting light at wavelengths from the ultraviolet to the visible.[1,2] In particular, InGaN multiple quantum wells (MQWs) are being used as the active layer of optical devices such as light emitting diodes[3] and laser diodes.[4–6] To realize devices with superior characteristics, quantum dot (QD) structures are desirable: a laser with QDs embedded in the active layer is expected to have lower threshold current and other superior characteristics, compared to QW lasers.[7] In InGaN QWs, it is said that nanometer-scale fluctuations of the indium content lead to "QD-like" states.[8,9] However, to realize the predicted performance of QD devices, an intentionally controlled QD structure is needed. Two methods have been used to fabricate InGaN QDs: self-assembled growth[10–13] and selective growth. Only a few studies have been conducted on the selective growth of InGaN.[14–17] The growth of pyramids of bulk InGaN on a GaN layer was demonstrated on a SiO_2/GaN/sapphire substrate patterned by conventional photolithography.[15] Also InGaN QD structures were formed on a Si/GaN/sapphire substrate patterned by focused ion beam etching, and the cathodoluminescence was investigated at 80 K for a few tens of QD structures.[17]

In this letter, we report fabrication of InGaN QD structures on a SiO_2/GaN/sapphire substrate patterned by conventional photolithography, using metalorganic chemical vapor deposition (MOCVD) with selective growth. InGaN structures were realized, with good uniformity across the sample, after optimizing the growth conditions. The photoluminescence (PL) spectrum shows a clear peak from InGaN QD structures at room temperature. To observe the emitting regions directly, we measured micro-PL intensity images with a spatial resolution of a few hundred nanometers. We observe the emission to come only from the tops of the hexagonal pyramid structures. The full width at half maximum (FWHM) of the spatial distribution of the intensity is 330 nm, which is comparable to the spatial resolution of the micro-PL intensity images. Such a narrow width of emission areas is supported by the fact that InGaN QDs are formed on the tops of pyramids.

The sample was grown using an atmospheric-pressure two-flow MOCVD system with a horizontal quartz reactor. After a 25 nm GaN nucleation layer was grown at 480 °C on a (0001)-oriented sapphire substrate, a 2-μm-thick GaN layer was grown at 1071 °C at a growth rate of 2.2 μm/h. During the growth of the GaN layer, the flow rate of trimethylgallium was 88 μmol/min with carrier gases of H_2 and N_2 at 4.0 and 11.5 slm, respectively. NH_3 was used as the group V source with a flow rate of 4.0 slm, which corresponds to a V/III ratio of about 2000. 40 nm of SiO_2 was deposited on the GaN by sputtering. The pattern was grid-like, with period 4 μm and square openings of side length 2 μm, and was prepared using conventional photolithography and a buffered HF solution. Selective growth of GaN was then performed, using atmospheric-pressure MOCVD again at a growth temperature of 945 °C. The flow rates of trimethylgallium TMG and NH_3 were 32 μmol/min and 4.0 slm, respectively. The flow rates of the H_2 and N_2 carrier gases were the same as before. Uniform hexagonal pyramids of GaN were realized, as confirmed by scanning electron microscopy (SEM). Selective growth of three periods of InGaN MQWs followed at 720 °C. The growth times for the InGaN well and $In_{0.02}Ga_{0.98}N$ barrier materials were such as give 2.4 and 4.1 nm thicknesses, respectively, in planar growth. Then, a 20-nm-thick $In_{0.02}Ga_{0.98}N$ layer was deposited. We consider that InGaN QD structures are formed at the tops of the hexagonal pyramids, as illustrated schematically in Fig. 1(a), as demonstrated in a GaAs system.[18]

Figure 1(b) shows a SEM bird's-eye-view of the final structures. As can be seen in Fig. 1(b), selective growth of InGaN occurred: no material was deposited on the SiO_2 mask. Figure 1(c) shows a cross-sectional image after the sample was cleaved. In Fig. 1(c), there is a pattern of horizontal streaks and the top of a hexagonal pyramid looks as if it was damaged in cleaving the sample. It is difficult to estimate the exact radius of curvature, however, the radius of curvature is no more than 30 nm. This indicates that very

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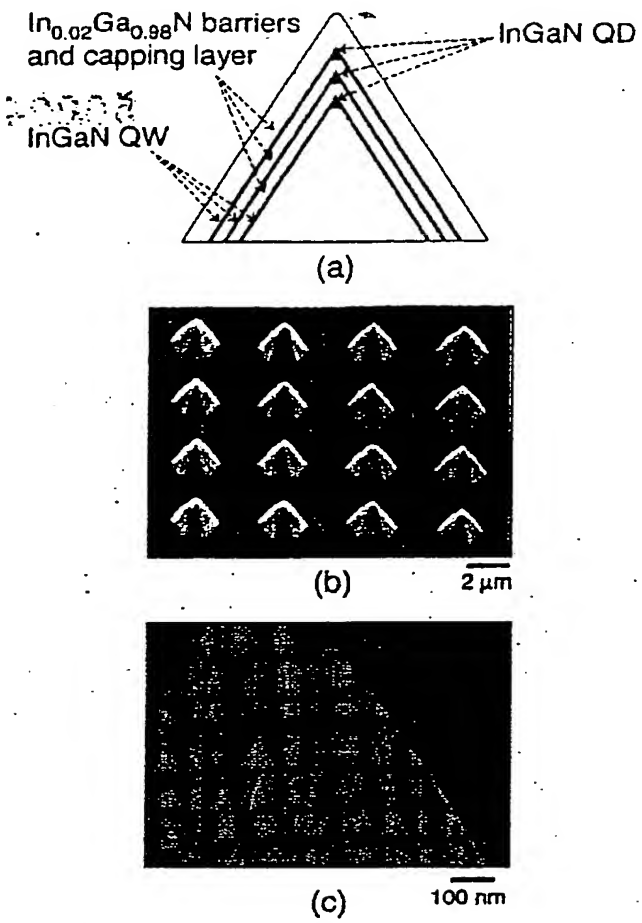

FIG. 1. (a) A schematic of InGaN QDs formed on the tops of hexagonal pyramids of GaN. SEM pictures of the sample: (b) bird's-eye-view and (c) cross section.

sharp tops were realized. The lateral size of InGaN QDs is considered to be comparable to the radius of curvature, no more than 30 nm, from the value obtained by cross-sectional SEM images. To discuss the lateral size of InGaN QDs more precisely, cross-sectional transmission electron microscopy would be help.

PL spectra were measured at room temperature. The excitation source was a He–Cd laser giving light with excitation energy of 3.82 eV and excitation power at the samples of 5 W/cm^2. The diameter of the laser spot on the sample was 0.7 mm, so about 25 000 QD structures were excited. The emission was dispersed by a 30 cm monochromator, and detected by a liquid nitrogen-cooled charge-coupled device (CCD) camera. Figure 2 shows the PL spectrum of the sample. As shown in Fig. 2, the peak energy of the emission attributed to the InGaN QD structures is 2.88 eV (430 nm), and the FWHM is 290 meV. The spectrum is broad because so many QD structures were excited. The intensity of the InGaN QD PL peak is smaller than that of the GaN emission peak at 3.42 eV, because the volume of InGaN QDs is much smaller than that of GaN bulk.

To identify the regions giving the emission at 430 nm, micro-PL intensity images were recorded at room temperature, using a conventional optical microscope equipped with

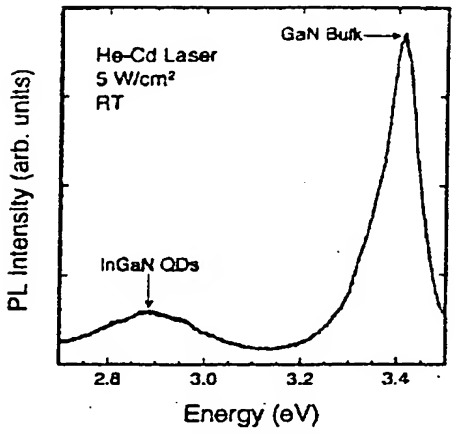

FIG. 2. Photoluminescence spectrum of the sample at room temperature.

an objective lens of which the nominal magnification factor was 100 and the numerical aperture (NA) was 1.3 when immersion oil was used, giving a typical spatial resolution as high as 150 nm.[19] The excitation source was a mercury lamp and the excitation wavelength could be selected using an excitation filter. The sample was uniformly excited and the collected light could be filtered by a high-pass barrier filter and a band-pass filter with a bandwidth of 5 nm, before being detected by an electrically cooled CCD camera with 1.3 million pixels. Figure 3(a) shows an image obtained only through a barrier filter of which the cutoff wavelength was 400 nm, without the excitation filter or band-pass filter, consisting mainly of reflected light. In Fig. 3(a), hexagonal shapes can be seen very clearly. This demonstrates that high spatial resolution is achieved. This is because we used an excitation source of short wavelength and an objectiv lens of NA as high as 1.3 with the immersion oil.

Figure 3(b) shows a micro-PL intensity image of PL of wavelength around 430 nm, accumulated over 10 s. The same area was observed in Fig. 3(b) as in Fig. 3(a). A 365 nm filter was used in excitation and excitation power at the sample was 1 W/cm^2. In collection, wavelengths shorter than 390 nm were cut off by a barrier filter and a 430 nm band-pass filter of bandwidth 5 nm was used, so the detected light was from the InGaN QD structures only, not from the GaN bulk or from the excitation source. The PL emission in Fig. 3(b) is only from the tops of the pyramids. In the cross-sectional profile of PL intensity in Fig. 3(c), the FWHM of 330 nm is comparable to the spatial resolution. Such a narrow width of emission areas indicates that the emission originates from InGaN QD structures embedded in the $In_{0.02}Ga_{0.98}N$ matrix.

We discuss the FWHM with respect to the micro-PL intensity image. As the PL was accumulated for 10 s, drift of the sample is not negligible. As the FWHM is comparable to the spatial resolution of the micro-PL intensity images, we cannot accurately compare the emission width from the micro-PL intensity images and the radius of curvature (no more than 30 nm) from the SEM images. Further investigation is needed t verify the three-dim nsional confin ment effect in detail, using magneto-PL or a measurement method with high spatial and spectral resolution, such as near-field scanning optical microscopy or cathodoluminescence. How-

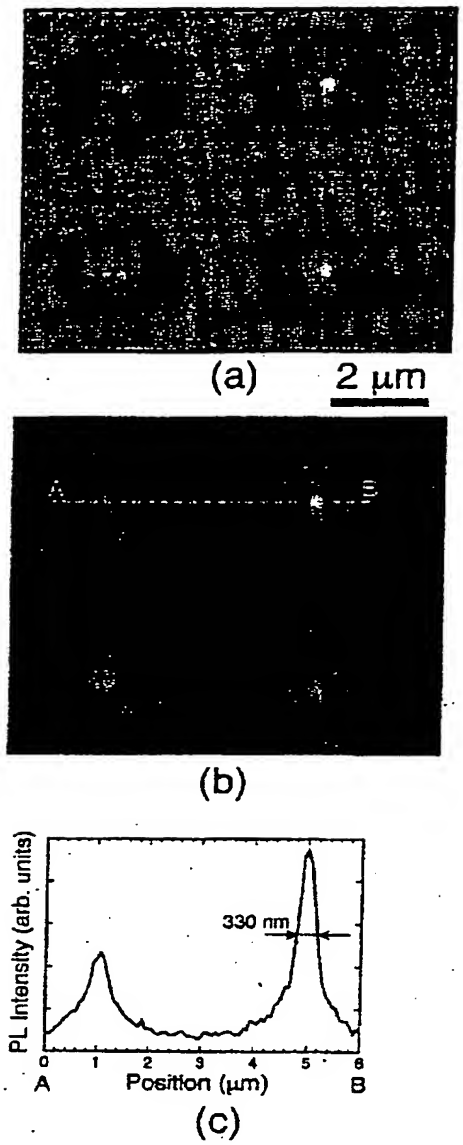

FIG. 3. (a) Reflective image, (b) micro-PL intensity image at a wavelength of 430 nm, and (c) cross-sectional profile of PL intensity along the line AB in (b).

ever, we have demonstrated in the micro-PL intensity images that the emission is only from the tops of the hexagonal pyramids, not from the edges.

In summary, we have fabricated InGaN QD structures, using selective MOCVD growth. By choosing the growth conditions carefully, a uniform array of hexagonal pyramids was realized. Intense PL was observed from the sample at room temperature. Micro-PL intensity images with a spatial resolution of a few hundred nanometers reveal that the emission was only from the tops of the pyramids. The spatial dimensions of the emitting area are comparable to the spatial resolution of the micro-PL intensity images. Such a narrow width of emission areas indicates the existence of InGaN QDs on the tops of the hexagonal pyramids.

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[1] S. Nakamura and G. Fasol, *The Blue Laser Diode* (Springer, Heidelberg, 1997).
[2] I. Akasaki and H. Amano, Jpn. J. Appl. Phys., Part 1 36, 5393 (1997).
[3] T. Mukai, M. Yamada, and S. Nakamura, Jpn. J. Appl. Phys., Part 1 38, 3976 (1999).
[4] S. Nakamura, M. Senoh, S. Nagahama, N. Iwasa, T. Yamada, T. Matsushita, H. Kiyoku, and Y. Sugimoto, Jpn. J. Appl. Phys., Part 2 35, L74 (1996).
[5] I. Akasaki, S. Sota, H. Sakai, T. Tanaka, M. Koike, and H. Amano, Electron. Lett. 32, 1105 (1996).
[6] S. Nakamura, M. Senoh, and S. Nagahama, T. Matsushita, H. Kiyoku, Y. Sugimoto, T. Kozaki, H. Umemoto, M. Sano, and T. Mukai, Jpn. J. Appl. Phys., Part 2 38, L226 (1999).
[7] Y. Arakawa and H. Sakaki, Appl. Phys. Lett. 40, 939 (1982).
[8] S. Chichibu, T. Azuhata, T. Sota, and S. Nakamura, Appl. Phys. Lett. 69, 4188 (1996).
[9] Y. Narukawa, Y. Kawakami, M. Funato, Sz. Fujita, Sg. Fujita, and S. Nakamura, Appl. Phys. Lett. 70, 981 (1997).
[10] H. Hirayama, S. Tanaka, P. Ramvall, and Y. Aoyagi, Appl. Phys. Lett. 72, 1736 (1998).
[11] K. Tachibana, T. Someya, and Y. Arakawa, Appl. Phys. Lett. 74, 383 (1999).
[12] K. Tachibana, T. Someya, Y. Arakawa, R. Werner, and A. Forchel, Appl. Phys. Lett. 75, 2605 (1999).
[13] C. Adelmann, J. Simon, N. T. Pelekanos, Y. Samson, G. Feuillet, and B. Daudin, Phys. Status Solidi A 176, 639 (1999).
[14] O. Gfrörer, J. Off, A. Sohmer, F. Scholz, and A. Hangleiter, Mater. Sci. Eng., B 50, 268 (1997).
[15] D. Kapolnek, S. Keller, R. D. Underwood, S. P. DenBaars, and U. K. Mishra, J. Cryst. Growth 189/190, 83 (1998).
[16] A. Inoue, T. Sakaguchi, Y. Moriguchi, M. Iwata, T. Miyamoto, F. Koyama, and K. Iga, *Proceeding 2nd International Symposium Blue Laser and Light Emitting Diodes*, 1998, p. 170.
[17] J. Wang, M. Nozaki, M. Lachab, Y. Ishikawa, R. S. Qhalid Fareed, T. Wang, M. Hao, and S. Sakai, Appl. Phys. Lett. 75, 950 (1999).
[18] Y. Nagamune, M. Nishioka, S. Tsukamoto, and Y. Arakawa, Appl. Phys. Lett. 64, 2495 (1994).
[19] T. Someya and Y. Arakawa, Jpn. J. Appl. Phys., Part 2 38, L1216 (1999).